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Modeling Ozone Concentrations: Urban- and Regional-Scale Models are Needed to Develop Emission Control Policies That Reduce Ozone Levels

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16. ABSTRACT

Regional scale air quality models are needed for coordinated planning of emission control strategies to reduce the ozone pollutant burden. Urban scale models by themselves cannot provide the link from one source area to the next. In this paper the structural components of the more rigorous grid-type regional ozone models are described along with some of the issues that must yet be resolved to achieve credible regional scale simulations. Chief among these issues are the questions of the magnitude and photochemical effects of regional-scale biogenic organic emissions, the mass transfer from the lower to upper boundary layer by cloud fluxes, the lack of determinism in regional scale wind fields, and perhaps most critically, how to properly describe the subgrid scale chemical and physical processes within the enormous volume of a regional grid cell. The major components and recent applications of five representative regional scale air quality models are presented.

17. KEY WORDS AND DOCUMENT ANALYSIS

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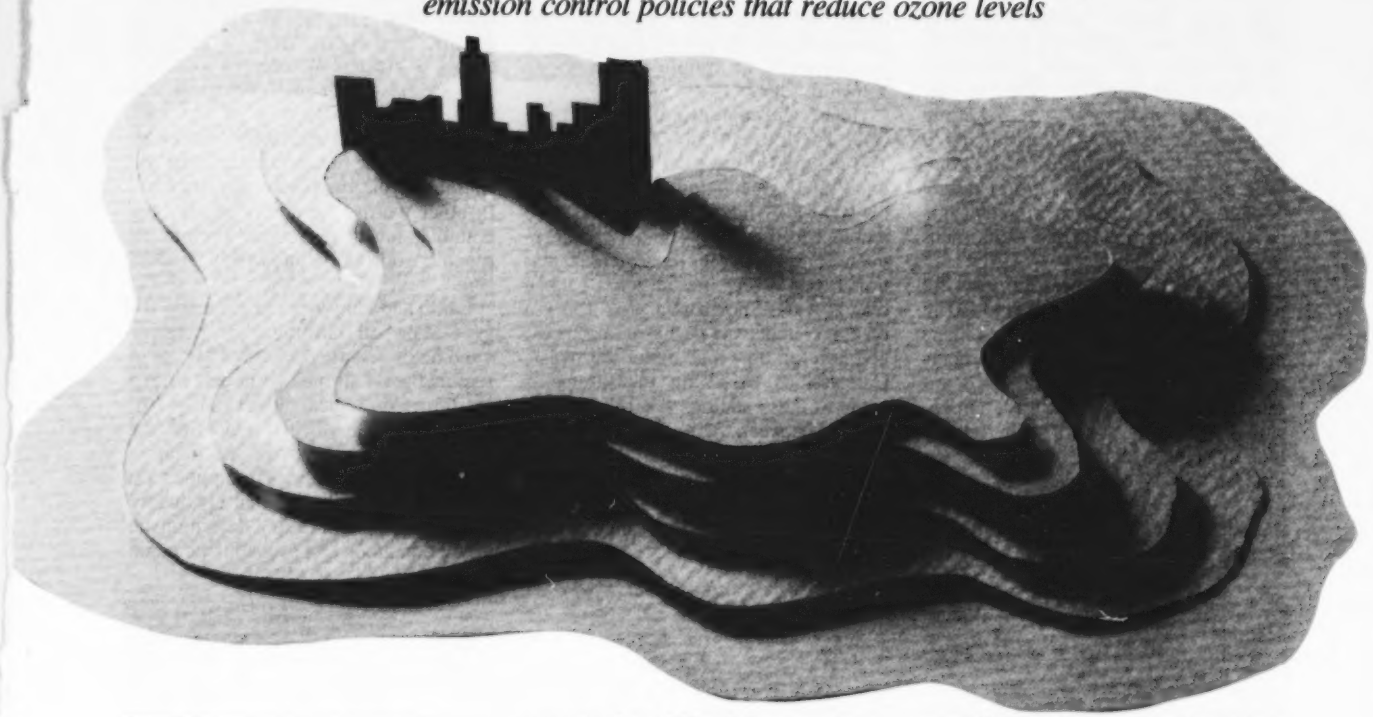
Modeling ozone concentrations

Urban- and regional-scale models are needed to develop emission control policies that reduce ozone levels



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Efforts to model ambient ozone (O_3) concentrations within urban areas have been under way for more than 15 years (*1–3*). Recorded events of O_3 and oxidant damage to materials and plants as well as its irritating effects on human tissues have been noted for an even longer period in areas affected by high ozone levels. During this time the realization arose that the urban O_3 problem often was a subcomponent of a larger, regional-scale ($\approx$ 1000 km) O_3 episode. The long-range transport of ozone between urban areas has become increasingly evident (*4–8*). As a result, model development programs to address the regional-scale O_3 issue began about 10 years ago and continue today.

Ozone not only affects health and welfare adversely in urban areas; it is becoming clear that O_3 damages crops and forest lands outside cities as well. Recent estimates of direct crop losses, based on the limited data available, suggest that about $3 billion would be lost annually if the United States experienced a seasonal daylight mean O_3 concentration of 60 ppb for seven hours each day (*9*). This value is within the range of frequently occurring daylight ozone concentrations in many agricultural areas of the United States.

The current primary National Ambient Air Quality Standard (NAAQS) for ozone, 120 ppb for one hour, is designed to protect human health. EPA is considering the issuance of a secondary standard to protect the nation's crops and forests from ozone damage. This standard is likely to specify a longer time period for estimating average ozone concentration, such as the 7-h seasonal daylight level. The cost of controlling pollution sources in order to bring areas that currently violate the ozone NAAQS into compliance is estimated to be in the billions of dollars. No estimates have yet been made of the control costs associated with a secondary ozone standard.

Only regional-scale air quality models for ozone can simulate concentrations over a sufficiently long time and wide area in a manner that is compatible with a secondary NAAQS standard. Such models are necessary components in the cost–benefit analysis that must be performed to assess the economic and social results of the emissions controls required to reduce ozone levels.

Meeting ozone reduction goals

Tropospheric ozone has been regulated since 1970, and nationwide levels have declined by about 10% between 1983 and 1984. Nevertheless, in the summer of 1986, EPA Administrator Lee Thomas announced that at least 28 metropolitan areas will have failed to meet the ozone concentration deadlines set for Dec. 31, 1987. By the end of 1987, this number turned out to exceed 50.

Technical issues

Determining wind flow patterns. Several technical problems, which are of minor significance in small-scale ozone models, must be considered for regional-scale ozone models. Foremost among them is the uncertainty in determining the correct wind flow pattern to use in the transport component of the model. The wind flow field may be obtained either by inference from meteorological observations or from the predictions of a meteorological model. The former method is more commonly

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used to determine wind flows for air quality models.

The construction of a wind flow field often is a problem for urban-scale models as well as for regional models. In urban areas there typically is an inadequate number of meteorological monitors to determine a well-resolved wind field. Special studies, on occasion, have been conducted in urban areas in which the regular monitoring network is enhanced by additional meteorological monitors. The resulting network is useful for constructing a wind field. On the regional scale, however, the spatial extent of the modeling domain is too large to augment the monitoring network in the same manner.

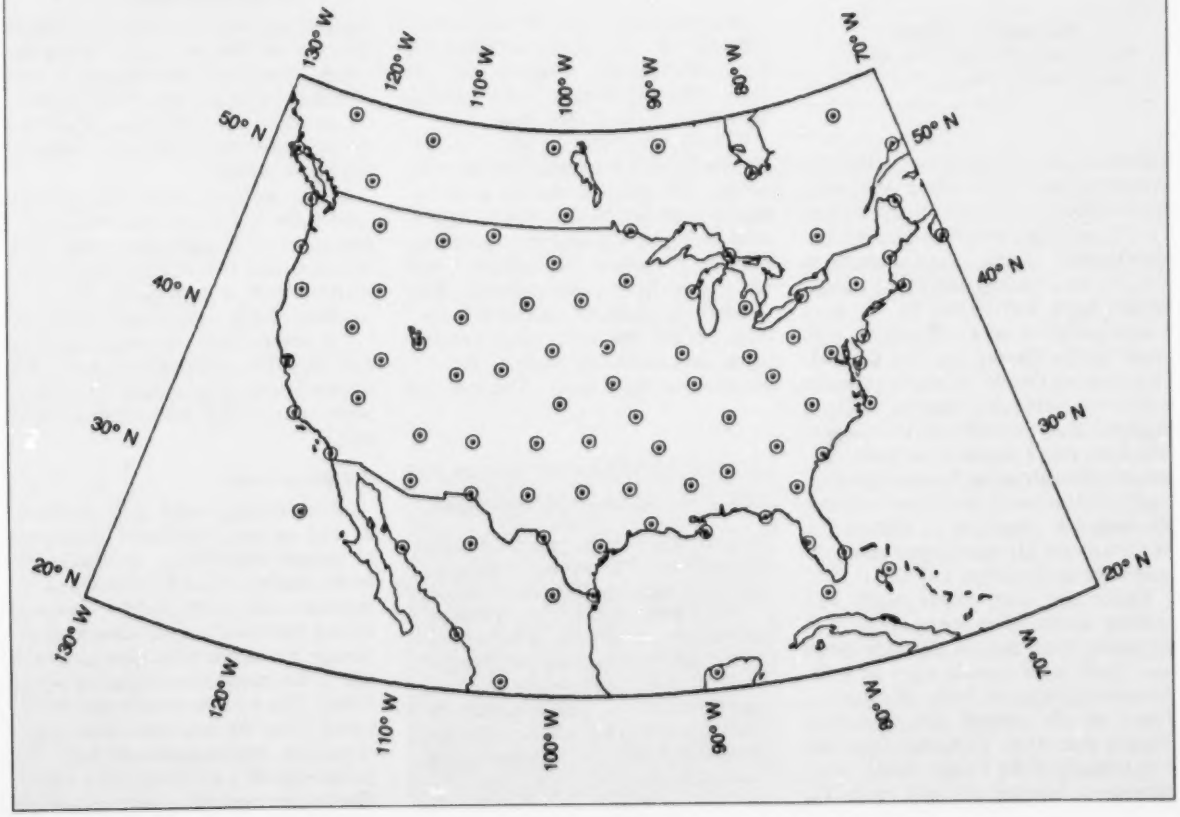
Two types of wind measurements are needed to form a wind flow field for an air quality model: measurements made near the surface and measurements made aloft. Both are necessary to determine the characteristics of three-dimensional transport for the air quality model. In most regions of North America, the density of surface wind monitors is relatively high. Surface wind measurements, however, are affected greatly by their immediate environment, so their ability to represent air flow patterns in a model is limited in spatial extent. Upper air wind measurements are more difficult to obtain than wind measurements at the surface, but they are applicable over a larger spatial area because the highly localized frictional effects of the earth's surface no longer are significant. Upper air wind measurements are made twice a day at the network of monitoring stations shown in Figure 1.

Upper air monitoring stations typically are located 300–500 km apart. The number of these stations used in a wind field calculation for a regional ($\approx$1000 km) model domain therefore would vary between 4 and 12. Consequently, one upper air measurement station would determine the wind pattern over a square horizontal area 300 km on a side under the best circumstances, and 500 km on a side under the worst. Calculations based on measurements made at these stations assume an even spacing between them. By comparison, the typical spatial resolution required in a regional air quality model varies from 20 to 100 km.

In any attempt to determine a spatially gridded wind field pattern from upper air observations, an inherent uncertainty resulting from the 300–500-km distances between stations is unavoidable. The uncertainty in the trajectory of an air parcel, determined from this wind field, may grow with time as its distance from the origination point increases. In this context, it becomes clear that the accuracy of air quality predictions from a regional model is greatest at receptor locations that are near both meteorological stations and major sources of pollutant emissions. This predictability decreases with increasing distance from either the sources or the meteorological stations. In other words, the ability to attribute pollutant levels in remote regions to specific sources is inherently limited (*9*).

Lamb and Hati propose a method for generating an ensemble of possible wind fields that are applicable for use in regional-scale air quality models from a set of meteorological observations (*10*). This concept explicitly addresses the uncertainty inherent in the specification of atmospheric states. The application of this concept for regional air quality models takes them from a deterministic prediction mode to a probabilistic mode

FIGURE 1
North American upper air wind-monitoring stations



in which the prediction of the pollutant concentration at a receptor is in the form of an expected probability distribution. Lamb plans to test these ideas with the EPA Regional Oxidant Model (ROM) in the future (*11–13*).

Cloud venting. Another issue of continuing interest in the regional-scale modeling of ozone is the process of cloud "venting," or the vertical transport of mixed-layer material aloft into the free troposphere by cumulus clouds formed by convection (*14*). This issue is not as relevant to smaller scale models in which material lost through the top of the mixed layer does not become reentrained into the model domain during the simulation. In a regional-scale model, however, material that is vented above the mixed layer can be transported considerable distances downwind by the faster-moving air flow in the free troposphere. This material then can be reentrained into the mixed layer far downwind from the original source area the following day as the growing mixed layer incorporates the air aloft. This process occurs well within the time and space scales of a regional model simulation.

The mechanism of cloud venting is described conceptually by Ching (*14*). The scenario begins in the morning when the growing layer of well-mixed air above the surface becomes photochemically active and its concentrations of O_3 increase rapidly relative to ozone concentrations found aloft. This mixed layer of air quickly deepens, and cumulus clouds begin to appear when the layer rises to a height equal to or greater than the convective condensation level. The cloud material is derived primarily from the mixed layer, and the developing cloud layer adjusts for this mass input of mixed-layer air by an appropriate amount of subsidence.

The mixed-layer ozone is carried aloft in the convective cells, and a net upward transport is possible when the mixed layer contains higher ozone concentrations than the cloud layer above. The significance of cloud venting for ozone therefore should increase with time during the afternoon. It also is possible that other atmospheric constituents are lifted through the cloud venting process. Of these, formaldehyde might be of particular significance because of its reactive potential and its higher concentrations during the afternoon. Occasional episodes of tall-stack emissions entrained directly into a strong convective cell also provide a means of injecting reactive species directly into the cloud layer.

Recent field experience has provided evidence that confirms the existence of the cloud-venting process and the return flow from cloud layer to mixed layer. In one experiment, a mass of nontoxic fluorescent dye particles was released below and above cumulus clouds while an airborne lidar monitored their transport and dispersion. Observations showed that the dye released below a cloud was vented upward into the cloud; the dye released at or above the top of a cloud rapidly descended into the mixed layer (*15*). In another experiment, airborne lidar measurements of O_3 and aerosols showed that deep convection can produce pollutant-rich elevated "reservoir" layers, which are transported over long distances without surface interactions (*16*). The experimental results suggest that approximately 30% of the maximum concentration of ozone measured at the surface in Norfolk, Va., on July 31, 1981, was caused by reactions of precursors that originated from external sources more than 500 km upwind.

Biogenic vs. anthropogenic emissions. With respect to source emissions, there still is substantial uncertainty about the effect of naturally occurring (biogenic) organic emissions on rural ozone concentrations. This issue needs to be explored further with regional-scale photochemical air quality models. Biogenic organic species, mostly isoprene and several monoterpenes, are emitted by many types of forest trees and cultivated plants. A number of techniques are available to determine the flux of these chemical species from individual plants or forest groves (*17, 18*), although there is substantial variation among measurements.

The emissions of isoprene generally increase with increases in ambient temperature and light intensity, and the emissions of monoterpenes increase with temperature. Altshuller summarizes the experimental results that explore the effects of temperature and light intensity on these emissions (*19*). There obviously are seasonal and geographical differences between the biogenic organics emission rates; maximum emission rates are observed during the warmer summer months in the heavily forested eastern and southern sections of the United States.

Biogenic emissions that are significant as compared with anthropogenic organic emissions in a particular localized area would be most likely to occur in rural areas of the Southeast during the summer. When the rates of these emissions are integrated over a large regional area ($\approx 1000 \times 1000$ km), however, the biogenic component can easily be the larger. In a recent analysis that used a biogenic emissions inventory compiled for the northeastern United States EPA-ROM application, it was found that the integrated anthropogenic organic emissions rate over the modeling domain was 1.7×10^7 kg/day; the total biogenic organic emissions rate was 2.6×10^7 kg/day for a mid-summer day with clear skies (*20*). These figures encompass an emissions area of approximately 1 million km^2, and they include nonreactive as well as reactive compounds. If the nonreactive compounds are excluded, the ratio of biogenic emissions to anthropogenic emissions drops to 1.2:1.

Individual field programs designed to measure ambient concentrations of biogenic organic species have shown that isoprene and α-pinene are the species most often identified (*19*). Outside forest canopies, the average concentrations of these species usually are less than 10 parts per billion of carbon (ppbC), although maximum isoprene concentrations of 30–40 ppbC were reported in several studies.

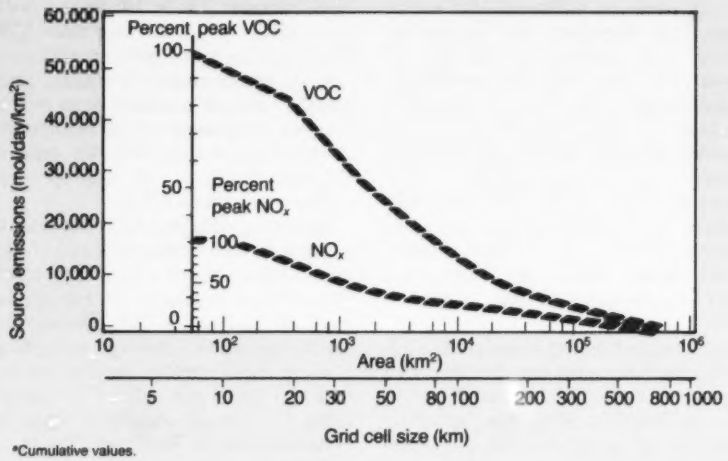
Measured concentrations of biogenic organic species often are lower than those expected from the estimates based on biogenic emission inventories. One explanation for this discrepancy may be the highly reactive nature of isoprene and the monoterpenes. Isoprene can contribute as much as 50% of the overall reactivity of rural air, even though its concentration is as low as 6% of the ambient hydrocarbon level (*21*). These species are rapidly oxidized into a number of intermediate carbon-containing species, such as methyl vinyl ketone, methylglyoxal, and formaldehyde, which also are highly reactive.

Altshuller notes that when the concentration ratios of isoprene to propene in forested areas are compared with regional average emission ratios, propene measurements are higher than expected (*19*). Propene, however, is a reactive hydrocarbon of anthropogenic origin and should form reactive products almost as fast as the isoprene. In addition, the average transport distance of the propene to a sampling point in a forested area must be substantially larger than that for isoprene. Regional oxidant models should be capable of helping resolve the apparent contradictions in the measurements and emission inventories of reactive biogenic species.

Several chemical kinetic mechanisms for the atmospheric reactions of isoprene recently have been postulated (*21, 22*). They are available for inclusion in photochemical air quality simulation models for applications in which an isoprene emissions inventory exists.

In an urban-scale modeling study, Lurmann et al. have applied a photochemical trajectory model to the Tampa–St. Petersburg, Fla., area to assess the role of biogenic organic emissions on urban O_3 formation (*23*). Al-

FIGURE 2
VOC and NO_x emissions vs. source area and characteristic length scale[a]



[a]Cumulative values.

though the biogenic and anthropogenic organic emission rates are estimated to be of comparable magnitude, the results of the study indicate that biogenic emissions contribute only 2–9% of the predicted maximum O_3 concentrations. The study also shows that this relatively small contribution is attributable to fast ozone–biogenic organic reactions that scavenge almost as much ozone as is produced by the biogenic organics in the urban environment.

On a regional scale, preliminary results from simulations of ozone transport in the northeastern United States with EPA's ROM model show noticeable impacts from biogenic emissions on ozone formation downwind of isolated major sources of nitrogen oxides (NO_x) in otherwise hydrocarbon-limited environments. The effect of biogenic emissions on ozone formation within most of the urban-source plumes in the ROM simulation were relatively small. Further studies of the effects of biogenic species in regional-scale applications are needed.

Major uncertainties also exist in the anthropogenic emission inventories used in regional models. Lamb explains that discrepancies between two recent emission inventories for the northeastern United States vary by a factor as large as 100 for emissions of NO_x in a given grid cell (*24*). Discrepancies between volatile organic carbon (VOC) emissions also are large. This noise in the emissions data can be larger than the changes in emissions that control strategies would cause.

This is a potential problem for regulators who use modeling scenarios as a partial basis for their decisions on emissions control measures, because the response of ozone concentrations to changes in VOC and NO_x emissions is a function of base emission levels. Therefore, if the estimated base emissions are erroneous, the simulated response of ozone concentrations to emission controls also will be erroneous. EPA and other groups responsible for compiling source emission inventories are working actively to reconcile discrepancies in the inventories and are producing more refined estimates of the true source strengths with each new generation of emission inventory.

The influence of stratospheric ozone. The intrusion of stratospheric ozone and its influence on concentrations of near-surface ozone is a frequently discussed issue that is beyond the range of investigation with regional air quality models. Generally, the upper boundary of the model domain is specified at a height varying from the top of the boundary layer (1–2 km above ground) to the mid-troposphere. Ozone concentrations above this level are specified as an upper boundary condition on the model and are determined by direct measurement (aircraft-based observations) or are inferred from ground observations.

Actually, large direct impacts of stratospheric ozone intrusions at ground level are unlikely. Analysis of the few published observations of high O_3 levels in stratospheric intrusions and of anomalously high O_3 at ground level attributed to stratospheric intrusions suggests that direct ground-level impacts may occur less than 1% of the time and usually are associated with ozone concentrations of 100 ppb or less (*25*). Regional models therefore will not conclusively quantify the near-surface impacts of stratospheric O_3.

Providing adequate spatial resolution. The final issue of regional ozone modeling encompasses the approximations made in solving the finite-difference analogs to the actual differential equations that describe over time the rates of changes in concentration of species that participate in chemical interactions in the model. This issue is important for most numerical air quality simulation models but has some special significance for regional-scale grid models. Modeling on this scale requires the model domain to cover a large horizontal area while holding the number of grid cells down to insure reasonable computation costs. This criterion is one factor that determines the spatial resolution achieved by the model; larger grid cells will provide coarse resolution and smaller cells will provide finer resolution.

Urban-scale ozone grid models typically use grid cells in a size range of 2–8 km to a side. This size generally is too small for regional models because an excessive number of grid cells would result. The problem, then, is to choose a grid cell size that will limit the overall number of cells and provide adequate spatial resolution.

How do we define adequate resolution in a regional ozone air quality model? We must be able to resolve the significant sources of emissions of ozone-forming species; otherwise, we risk diluting them into a large volume in which the model is likely to diminish their significance and underpredict the resulting O_3 concentrations.

Using the National Acid Precipitation Assessment Program emissions inventory (*24, 26*), Lamb has studied the issue of "scale length" of significant nationwide sources of VOC and NO_x emissions, the primary precursors of ozone in the northeastern United States. Following his analysis, let us rank the counties with the strongest emission fluxes (mol km^{-2} day^{-1}) in the study area from highest (No. 1) to lowest. Now let us define a cumulative emissions function, Q, that represents the area-weighted average of the individual county emissions from the greatest down to and including county No. N from our list:

$$Q = \sum_{i=1}^{N} Q(i)A(i) \Big/ \sum_{i=1}^{N} A(i)$$

where $A(i)$ is the area of county i. A plot of the cumulative emissions function, Q, vs. the cumulative area is shown in Figure 2 for both VOC and NO_x. The left edge of the upper curve corresponds to the highest VOC emissions source county, Q_{VOC} = 53,457 mol km^{-2} day^{-1} and A = 58.1 km^2. In order to define this strongest source,

we would need a model grid size of $A^{1/2}$, or 7.6 km. In other words, a model with grid size of 7.6 km would define 100% of the peak source strength.

If we follow the curves, we see that a grid size of 20 km would determine about 80% of the peak VOC and NO_x source strengths, provided that the strongest VOC and NO_x sources fall within the same grid cells. Likewise, a grid size of 50 km would resolve slightly less than 50% of the peak source strength. A model with grid cells of 500 km is essentially no better than one in which the entire domain is treated as a single cell. When a model is unable to resolve the strengths of the actual sources, errors will occur in the simulated concentrations of photochemical species. Recent applications of regional O_3 air quality models have used grid sizes from 18 to 100 km.

The method of numerical solution of chemical kinetics equations is another area in which approximations made to achieve a faster solution may produce errors in the model results. The kinetics modules of a photochemical air quality model typically consume 75–90% of the total model execution time and thus are prime candidates for efforts to achieve greater solution efficiency. Problems associated with solving the chemical equations arise because the time scales of the various reactions composing the kinetic mechanism span a range of several orders of magnitude. This makes it difficult to choose an optimum time increment for the solution. Such a set of reactions is characterized as "stiff."

Accurate numerical solvers based on the Gear method (*27*) usually are successful for stiff systems but consume large amounts of computer time and memory. This makes them inappropriate for use in large grid models of air quality simulation, although they may be tractable in trajectory or box models. The more common method of solution in grid models uses a more approximate method based on steady-state chemistry or "quasi-steady-states" that compares well with Gear-type solutions when all empirical parameters, such as the solution time increment, are optimally adjusted for accurate solutions over the range of expected ambient species concentrations (*28*). Outside the range of the optimally adjusted parameters, however, the solutions can degrade quickly.

Regional models for ozone

The air quality simulation model attempts to provide a mathematical solution to the atmospheric diffusion equation for all species of interest. Generally, analytic solutions for the set of model equations are not available. Iterative numerical solutions to the finite-difference analogs of the original differential equations therefore are used. The terms in the model equations include those for three-dimensional transport and diffusion, pollutant sources and sinks, and chemical transformations. The framework of the model in which these equations are solved can vary from a three-dimensional Eulerian grid model to a moving trajectory model to the relatively simple box model. Hybrid formulations between the three types of models also are possible. In our discussion of regional ozone modeling, we limit ourselves to the grid models. Most of its concepts apply to the other model types, however, since their formulations essentially are subsets of the grid model formulation.

All grid models include representations of the advective transport terms. The transport considers the movement of mass as governed by the mean wind velocity. (In regional-scale O_3 models, mean quantities generally are averaged over a period of 1–3 h.) Models vary in the methods by which the gridded three-dimensional wind field is specified. The most common device is the use of an objective analysis technique to interpolate between observations of wind speed and direction (*29*). The interpolation technique sometimes is enhanced by mass-conservation principles to improve the results in areas of complex flow such as those with sea breeze circulations or mountain–valley systems (*30*).

In the most rigorous approach, a dy-

Ozone measurement methods in current use

Several techniques for measuring ozone concentration have gained wide acceptance. EPA's standard reference method is the ozone–ethylene chemiluminescence method, by which a sample of ambient air that contains ozone is mixed with pure ethylene that flows at a slow, controlled rate. One product of the resulting reaction is formaldehyde in an excited state, which can be detected by a photomultiplier tube. This technique is ozone-specific and has a minimum detection limit of about 5 ppb and a response time of less than 30s. A number of commercially available instruments use this method.

A related technique, less commonly used, also is based on the chemiluminescence principle. With this method, ozone reacts with solid rhodamine B. Although this technique has a slightly longer response time, its minimum detection limit is 1 ppb.

Another technique is based on ultraviolet (UV) photometry. The degree of UV absorption is detected within a particular wavelength band in which ozone is absorbed. This technique is ozone-specific and has a response time of about 30 s and a minimum detection limit of about 5 ppb. It is used by some commercially available instruments and often serves as a calibration reference for chemiluminescence instruments.

Continuous ozone-monitoring instruments in current use are quite reliable. Performance specifications for the instruments show precision values within 10 ppb. In continuous field operation, the expected variability or error in a measurement may be 10–30%.

Most of the O_3 data available in the United States today from regular continuous-monitoring stations have been collected at urban or suburban sites; a typical urban area contains one to five monitors. Some of the larger metropolitan urban areas contain more monitoring sites. These data are compatible with the urban model simulation studies in which the measured O_3 concentrations are used to define initial or boundary conditions and to evaluate the models.

Although it is sufficient for most urban model application studies, the existing O_3 data base is far from ideal in terms of the number and spatial distribution of monitors in metropolitan areas. For regional-scale modeling, the existing network is clearly insufficient because only a few monitoring stations are located in rural and "regionally representative" locations away from the influence of urban emission sources. Ozone monitors sometimes are located in rural areas for a limited amount of time during special field studies in a given region.

Because of concerns about O_3 damage to crops and forests, additional monitors are being installed in rural and remote areas on a permanent basis. Even in the most optimistic of futures, however, it would be unrealistic to expect a dense network of O_3 monitors on a regional ($\approx$ 1000 km) scale. Nevertheless, an increase in the number of monitors and a better spatial distribution of these monitors in the ambient O_3 data base will make the use of these data in regional-scale modeling applications easier and more meaningful.

namic wind field model calculates the gridded winds based on a numerical solution of the equations of mass, momentum, energy, and moisture (*31*). Once the wind field is determined, various numerical solution techniques may be used to solve the advective transport equation (*32*). Each technique has its own behavioral characteristics; some prevent negative concentrations from being calculated and others preserve the peak concentration areas better. The magnitude of the gradients in the spatial patterns of predicted concentrations is determined, in part, by the choice of transport solution technique.

Turbulent diffusion is the process whereby mass is spread out and diluted based on the perturbations of the wind field from the mean flow. This part of the flow field is not resolved on the time or space scales of the model. Typically this component is modeled empirically by the K-theory, or gradient transfer theory approach. This theory assumes that the local concentration of mass will change with time as a function of the mean wind speed, the local concentration gradient in space, and a stability-dependent empirical diffusivity coefficient. This approach also is convenient for numerically solving the diffusion component along with the transport.

Other methods of treating turbulent diffusion in regional models are possible, including a more rigorous approach to specifying the turbulent fluctuations themselves. If this approach is used, it usually is confined to the vertical direction, where the diffusive effects are closer in scale to the mean transport and thus have a greater relative impact than in the horizontal direction.

Modeling the kinetics of the relevant atmospheric photochemistry is an integral part of all regional-scale ozone models. It is difficult to condense the hundreds of tropospheric photochemical reactions that involve NO_x, oxygen (O_x), and hydrocarbon (HC) species into a set that can be simulated efficiently within an air quality model. The techniques for accomplishing this are varied and continue to evolve.

Currently, there are three generally accepted techniques:

- the surrogate species technique, in which the reactions of a few hydrocarbon species represent the entire atmospheric mixture of hydrocarbons (for example, the Dodge mechanism [*33*]);
- the lumped molecule technique, in which one hydrocarbon species, sometimes a generalized species, serves as a surrogate for a group of similarly reacting species (for example, the Carter–Atkinson–Lurmann–Lloyd (CALL) mechanism [*34*]); and
- the lumped structure, or carbon-bond method, in which hydrocarbon groupings are based on structural units (for example, the CBM-IV mechanism [*35*]).

Chemical mechanisms applicable to regional O_3 modeling must include the slower reacting compounds because of the multiday nature of the model simulations and certain reactive biogenic hydrocarbon species that may become important in rural areas. These chemical aspects have been ignored in urban-scale ozone models because their scope is more limited in time and space.

Our understanding of atmospheric chemistry of the O_3-NO_x-HC cycle has improved greatly during the past 20 years. Uncertainties still exist, however, and are reflected in all of the contemporary literature on chemical mechanisms. First of all, some of the reaction rate constants are highly speculative. Also, for regional-scale modeling, the more significant uncertainties include those associated with nighttime NO_x chemistry, the fate of many slower-reacting organic species, and numerous heterogeneous processes involving aerosols that generally are neglected in the examination of chemical mechanisms for the production of ozone. The degree of error introduced into mechanism predictions from these uncertainties has not been quantified adequately.

Most ozone predictions based on current knowledge of the applicable mech-

TABLE 1
Comparison of two regional air quality simulation models[a]

	Regional Oxidant Model (ROM)	Regional Transport Model (RTM-III)
Model type	Regional ozone	Regional ozone
Developers	EPA Atmospheric Sciences Research Laboratory, Research Triangle Park, N.C. (Robert Lamb)	Systems Applications, Inc., San Rafael, Calif. (Mei-Kao Liu and Ralph Morris)
References	*11–13*	*36, 37*
Horizontal grid resolution	18.5-km grid cells (780 × 1100-km model domain)	Variable, 10–80-km grid cells (1800 × 2000-km model domain in eastern U.S. application)
Vertical grid resolution	Three layers extending to height of tops of cumulus clouds and one diagnostic surface layer	Three layers extending to 2000–6000 m above ground level and one diagnostic surface layer
Temporal resolution	1 hour	1–3 hours
Subgrid-scale features	Parameterized estimate of near-surface concentration variance of NO, NO_2, and O_3 within a grid cell, based on heterogeneous emissions distribution	Gaussian-type point source module with O_3–NO_x photostationary chemistry at receptor points or, optionally, reactive plume module for point sources
Horizontal transport	Winds interpolated from observations, except nighttime jet-layer winds, which are derived from hydrostatic model. "Probabilistic" generation of wind fields. Numerical advection by biquintic[b] polynomial scheme	Winds interpolated from observations or generated by dynamic wind model. Numerical advection by Flux-Corrected Transport method (SHASTA or multidimensional FCT)
Chemical kinetics	Carbon-bond IV (28 species, 70 reactions). No steady-state approximations made. All species advected	Carbon-bond IV (28 species, 70 reactions). Solution procedure uses local steady-state approximations. Fourteen species advected
Deposition	Dry deposition parameterized by empirical relationship with friction velocity, Obukhov length[c], and surface resistance	Dry deposition parameterized with aerodynamic resistance of the laminar layer and surface resistance
Cloud processes	Direct cumulus cloud mass flux from surface layer to cloud layer	Cloud mass flux parameterization planned for the future
Applications	Northeastern United States (NEROS region); southeastern United States	Eastern United States (SURE region); Midwest–Southeast United States; California Central Valley; western Europe (PHOXA)

[a]3-dimensional grid type.
[b]2-dimensional, fifth order.
[c]Obukhov length is a characteristic scale length based on stability of the lower atmosphere.

anisms are quite similar for the same set of operating conditions. Fractional changes of some key parameters, however, may cause dissimilar responses among these mechanisms. This aspect disturbs policy-makers, who base their decisions, in part, on the response of ozone production mechanisms to fractional changes in concentrations of precursor species. Further development and understanding of these mechanisms and of all important chemical processes are needed before these differences can be reconciled.

Sources and sinks of pollutants are another essential aspect of regional models. Mass is removed either chemically (through the chemical kinetic mechanism), by rainout and washout (usually not considered in models for O_3, but important factors in acid deposition models), or by deposition to the surface. The latter process usually is characterized by a "deposition velocity" for a chemical species that is empirically linked to the resistance of the atmosphere as well as the resistance of the underlying surface to the downward movement of the species near the surface.

Pollutants typically include NO_x, HC, and carbon monoxide (CO) species from distributed-area and large point sources. The area sources are specified as emission fluxes averaged over the cross-sectional area of each grid cell. Point source emissions are added to the appropriate grid cell after any buoyant plume rise is accounted for. Some models attempt to treat the point source plume separately while it is of a subgrid scale size. Treating the subgrid scale effects, in one form or another, is a common issue in regional models, where a single source or an urban area occupies only a portion of a single grid cell.

Table I compares the characteristics of two currently used grid models for regional-scale air pollution. They are designed exclusively for the prediction of ozone concentrations as well as for those of the other constituent species of photochemical smog. Three other regional models have been designed for the simulation of the acid deposition phenomenon—the Sulfur Transport Eulerian Model (*38–40*), the Regional Acid Deposition Model (*41–43*), and the Acid Deposition and Oxidant Model (*44*). All of the models are operational, although development continues on each.

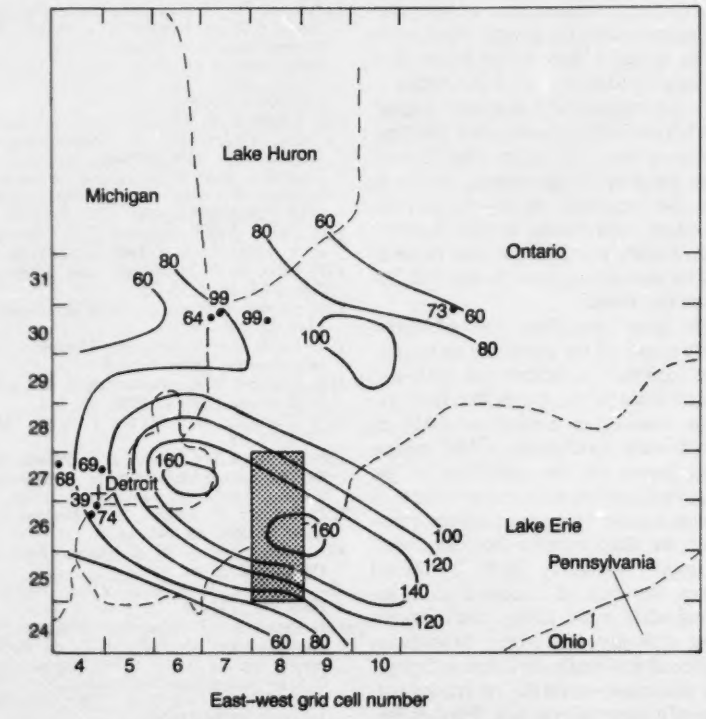
Models for the rigorous treatment of acid deposition must include all of the physical processes that affect photochemical smog as well as many others that uniquely affect acid deposition, such as in-cloud physical processes, rainout, washout, wet deposition, aqueous-phase chemistry, and others. Because O_3, the hydroxyl radical (OH), and hydrogen peroxide (H_2O_2) represent important oxidizing pathways in the chemistry of acid deposition (*45*), these models must simulate their concentrations accurately and include the primary precursor species in their emission inventories. Thus regional ozone concentrations are derived, albeit as a by-product, from regional-scale models of acid deposition.

Certain factors, such as spatial scales, separate the two model types. The overall domains and grid sizes of acid deposition models tend to be larger than those for regional ozone and may be too coarse for proper resolution of high-concentration areas. On the other hand, acid deposition models have more vertical resolution than ozone models to account for the physical processes in clouds properly.

Applications of the regional air pollution models so far have centered on the eastern parts of the United States and North America where multiday, multicity pollution episodes are most common and for which comprehensive data bases have been collected. Other areas of application, however, are being planned or are in progress. Particular interest in regional models exists in Europe because of the large-scale pollution episodes that can develop there and cross political boundaries. The governments of West Germany and the Netherlands are sponsoring development and application projects using the Regional Transport Model (RTM-III) and the Acid Deposition and Oxidant Model (ADOM).

Figure 3 illustrates one of the primary benefits of the use of regional oxidant models: defining individual urban plumes within the model domain. In a test application of the first generation ROM, maximum predicted O_3 concentrations were contoured over an area downwind of the Detroit–Windsor metropolitan plume during the second day of simulation of a pollution episode in August 1979. (Only a limited portion of the ROM grid is shown.) Although the principal axis of the plume extended out over Lake Erie, predicted ozone concentrations agreed fairly well with the pattern of surface measurements shown. The shaded region in the figure corresponds to an area of aircraft monitoring on Aug. 4, 1979, when the O_3 measurements aloft (within the well-

FIGURE 3
Contours of predicted maximum ozone concentrations [a]



[a]From ROM for the Detroit metropolitan plume, Aug. 4, 1979. Points are maximum observed concentrations at monitoring stations. Shaded area indicates grid cells in which measurements of ozone taken from aircraft were above the regional background value and as high as 150 ppb.

mixed layer) were observed to be as high as 150 ppb close to the area in which a concentration of 160 ppb had been predicted (*46*).

Issues to be resolved

There is considerable interest in regional-scale air quality modeling for O_3, other photochemical oxidants, and acid deposition. This interest is fueled by members of the scientific community and by policy-makers who now recognize that large-scale episodes of air pollution can span many days and cross many political boundaries. Coordinated planning of optimum emission control strategies to reduce the pollutant burden must be organized around a comprehensive approach to the problem. Part of the process includes the use of both regional-scale and urban-scale air quality models. Urban-scale models alone cannot provide the link between source areas as effectively as regional models.

The challenge to air quality regulators is to integrate the use of regional O_3 models with the urban models in a way that allows interaction between the different spatial scales and makes best use of the data provided by each type of model. The coarser resolution of regional models helps define the broad spatial patterns of ozone and the directional movement of these patterns over time; the finer scale of the urban models better locates areas of high concentration within an airshed. Nesting an urban model within the structure of a regional model for particular cities is one way to provide a dynamic interaction between the spatial scales. Another approach would be to maintain the separate integrity of the models, but to allow the regional model to provide boundary conditions to the smaller scale model, using the models in an iterative manner to provide the link between the scales.

We have described the structural components of the more rigorous grid-type regional O_3 models and have discussed some of the issues that have yet to be resolved to achieve credible regional-scale simulations. Chief among these issues are the questions of the magnitude and photochemical effects of regional-scale biogenic organic emissions, the mass transfer from the lower to upper boundary layer by cloud fluxes, the lack of determinism in regional-scale wind fields, and perhaps most critically, the proper description of the subgrid-scale chemical and physical processes within the enormous volume of a regional grid cell. Further theoretical research and field experiments will help provide the knowledge necessary to resolve these outstanding issues of regional-scale air quality modeling.

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